Global distribution and sources of volatile and nonvolatile aerosol in the remote troposphere

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[1] Airborne measurements of aerosol (condensation nuclei, CN) and selected trace gases made over areas of the North Atlantic Ocean during Subsonic Assessment (SASS) Ozone and Nitrogen Oxide Experiment (SONEX) (October/November 1997), the south tropical Pacific Ocean during Pacific Exploratory Mission (PEM)-Tropics A (September/October 1996), and PEM-Tropics B (March/April 1999) have been analyzed. The emphasis is on interpreting variations in the number densities of fine (>17 nm) and ultrafine (>8 nm) aerosol in the upper troposphere (8–12 km). These data suggest that large number densities of highly volatile CN ($10^4 - 10^5$ cm^{-3}) are present in the upper troposphere and particularly over the tropical/subtropical region. CN number densities in all regions are largest when the atmosphere is devoid of nonvolatile particles. Through marine convection and long-distance horizontal transport, volatile CN originating from the tropical/subtropical regions can frequently impact the abundance of aerosol in the middle and upper troposphere at mid to high latitudes. Nonvolatile aerosols behave in a manner similar to tracers of combustion (CO) and photochemical pollution (peroxyacetylnitrate (PAN)), implying a continental pollution source from industrial emissions or biomass burning. In the upper troposphere we find that volatile and nonvolatile aerosol number densities are inversely correlated. Results from an aerosol microphysical model suggest that the coagulation of fine volatile particles with fewer but larger nonvolatile particles, of principally anthropogenic origin, is one possible explanation for this relationship. In some instances the larger nonvolatile particles may also directly remove precursors (e.g., H_2SO_4) and effectively stop nucleation.   *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; 4801 Oceanography: Biological and Chemical: Aerosols (0305); *KEYWORDS:* aerosol, particles, upper troposphere, pollution, condensation nuclei, PAN

1. Introduction

[2] Aerosol particles of myriad composition with sizes ranging from a few nanometers to a few hundred microns can have an important impact on the chemistry and climate of the atmosphere [*Andreae and Crutzen*, 1997; *Warneck*, 2000]. These aerosol can originate from a variety of primary and secondary sources of natural and anthropogenic origin. A topic of current interest in the study of aerosol effects on climate is their formation and removal in the remote troposphere. Limited investigations in the Pacific region, North America, and western Europe sug-

gest an upper tropospheric source of fine particles [*Clarke*, 1993; *Hofmann*, 1993; *Schröder and Ström*, 1997; *de Reus et al.*, 1998; *Nyeki et al.*, 1999]. Injections from aircraft emissions also have been implicated as a substantial anthropogenic source of fine particles in the upper troposphere (UT) [*Schlager et al.*, 1997; *Hofmann et al.*, 1998; *Anderson et al.*, 1999; *Ferry et al.*, 1999; *Paladino et al.*, 2000]. The exact mechanisms by which these particles are formed and removed as well as their role in atmospheric radiative effects are not well understood.

[3] For a number of years, we have been investigating the influence of anthropogenic emissions on the composition and chemistry of the remote troposphere using instrumented aircraft. These investigations primarily have employed a NASA DC-8 aircraft to measure trace gases and aerosol between 0 and 12 km. In recent years, we have focused on the North Atlantic (Subsonic Assessment (SASS) Ozone and Nitrogen Oxide Experiment (SONEX), October/November 1997) and tropical South Pacific regions (Pacific Exploratory Mission (PEM)-Tropics A, September/October 1996; PEM-Tropics B, March/April

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1999). Overviews of these three missions including objectives, payload, analytical techniques, flight profiles, and meteorological characteristics are available elsewhere [*Hoell et al.*, 1999; *Singh et al.*, 1999; *Schumann et al.*, 2000; *Raper et al.*, 2001]. These field studies provided an opportunity to collect a unique tropospheric database on the concentration of particles and a vast array of gaseous chemical tracers of continental and marine origin (e.g., CO, peroxyacetylnitrate (PAN), dimethyl sulfide (DMS)). The tropospheric particle data were collected using three parallel TSI condensation nuclei (CN) counters with a 1-s time response described in more detail by *Cofer et al.* [1998]. These provided measurements of ultrafine CN (>8 nm in diameter, UFCN) and fine CN (>17 nm in diameter, FCN). Particles >1000 nm in diameter, expected to be extremely few in number, were not sampled during these measurements. These upper and lower cutoffs were determined based on a number of laboratory tests (B. Anderson, NASA Langley, private communication, 2001). The nonvolatile portion of FCN was determined by heating the sampled air to 300°C, which evaporated off nearly all sulfate and nitrate aerosol. In this paper we analyze the distribution and behavior of CN along with those of gaseous tracers, whose sources are better known. Our objective is to assess aerosol abundance, formation, and loss with a focus on the upper troposphere (8–12 km).

2. Atmospheric Vertical and Latitudinal Structure

[4] The vertical distributions of ultrafine, fine, and nonvolatile FCN number densities obtained in the troposphere over the North Atlantic (45–70°N) during SONEX, and the South Pacific (0–30°S) during PEM-Tropics A and PEM-Tropics B are shown in Figures 1a, 1c, and 1e. The corresponding nonvolatile fraction (NV-FCN/FCN) is plotted in Figures 1b, 1d, and 1f. The selected latitudes represent regions from where most of the data were collected and form a robust subset. All reported CN data were normalized to 20°C and 1 atm pressure. The total CN number densities increase with altitude with the maximum encountered in the UT. The greatest mean densities ($\approx 10^4$ cm^{-3}) were found in the tropical Pacific UT (Figure 1e). The UFCN number densities were generally twice as large as FCN (Figure 1), suggesting that a substantial fraction of the particles ($\approx$50%) were in the 8–17 nm range. There were individual events when this fraction was nearly 80%. The nonvolatile count was much larger in the NH (Figure 1a) compared to the SH (Figures 1c and 1e). During PEM-Tropics B, when the air was the cleanest, the nonvolatile FCN fraction was <10% in much of the troposphere (Figure 1f). This fraction was much higher over the North Atlantic, and most of the FCN in the lowest troposphere were nonvolatile (Figure 1b).

[5] Figures 2a and 2c present the mean latitudinal distributions (5° bands) of total and nonvolatile FCN in the UT (8–12 km) using data from SONEX (Atlantic) and PEM-Tropics B (Pacific). The corresponding profiles for select gaseous species (NO$_x$, CO, and PAN) are shown in Figures 2b and 2d. Both CO and PAN are excellent tracers of combustion and photochemistry, while NO$_x$ in the UT often indicates a tropospheric source (e.g., lightning, aircraft

emissions). Over both the Atlantic and the Pacific, fine CN number densities increased toward the tropics reaching a peak value of $5-10 \times 10^3$ cm^{-3} in the equatorial regions. The latitudinal behavior of nonvolatile particles, with lowest concentrations in the tropics, was opposite to that of FCN.

[6] The FCN (and UFCN) counts over the subtropical (20–30°N) North Atlantic (Figure 2a) were greater than those over the corresponding subtropical eastern Pacific (Figure 2c). We note that the high-latitude UT region of the North Atlantic (Figure 2a) can be influenced by direct fine particle emissions from subsonic aircraft [*Anderson et al.*, 1999]. This is somewhat corroborated by the elevation in NO$_x$ mixing ratios observed near 50°N (Figure 2b) which were associated with jet engine exhaust [*Singh et al.*, 1999]. Volatile and nonvolatile FCN counts also varied greatly in the Southern Hemisphere (SH) during the PEM-Tropics A and PEM-Tropics B austral seasons (Figure 1). The reasons for these differences will be discussed in section 3.1, but we note here that the SH atmosphere during PEM-Tropics A was far more impacted by pollution than during PEM-Tropics B. A comparison of mixing ratios for select primary and secondary gaseous tracers of pollution (e.g., CO, PAN) from the region 0–30°S shown in Figure 3 provides ample evidence for the presence of significant pollution during PEM-Tropics A. Extensive trajectory analysis and study of relationships between various tracers indicates that during PEM-Tropics A biomass burning pollution from South America and Africa had been transported to this region some 10^4 km away from its source [*Board et al.*, 1999; *Singh et al.*, 2000]. Conversely, the direct impact of continental pollution in the SH during the PEM-Tropics B was minimal [*Fuelberg et al.*, 2001].

3. Results and Discussion

[7] Particle and gas data have been analyzed from the North Atlantic and the South Pacific with a focus on the upper troposphere (UT). The variabilities in these particle mixing ratios are discussed in light of their sources, sinks, and transport.

3.1. Volatile and Nonvolatile Particles

[8] Volatile and nonvolatile particle counts from the UT (8–12 km) for each of the three missions are plotted in Figure 4 as a function of the NV-FCN fraction. This fraction would be 0 if all fine particles were volatile at 300°C and 1 if none were volatile. Typically, the only particles in the atmosphere that are still nonvolatile at 300°C are soot, soil dust, sea salt, and some biomass originated particles. The corresponding data for CO and PAN, both tracers of pollution, also are shown. Figure 4 indicates that in nearly all cases increasing nonvolatile fraction is not only associated with increasing nonvolatile counts but also with decreasing volatile (and total) counts. As is indicated by PAN and CO mixing ratios, high concentrations of nonvolatile FCN are nearly always associated with pollution signatures. Over the North Atlantic, nearly 70% of particles associated with events of CO > 100 ppb were nonvolatile. Five-day backward trajectories [*Fuelberg et al.*, 2000] indicated that these air parcels were transported across the Arctic and the North American continent from as far as Alaska and the North Pacific.
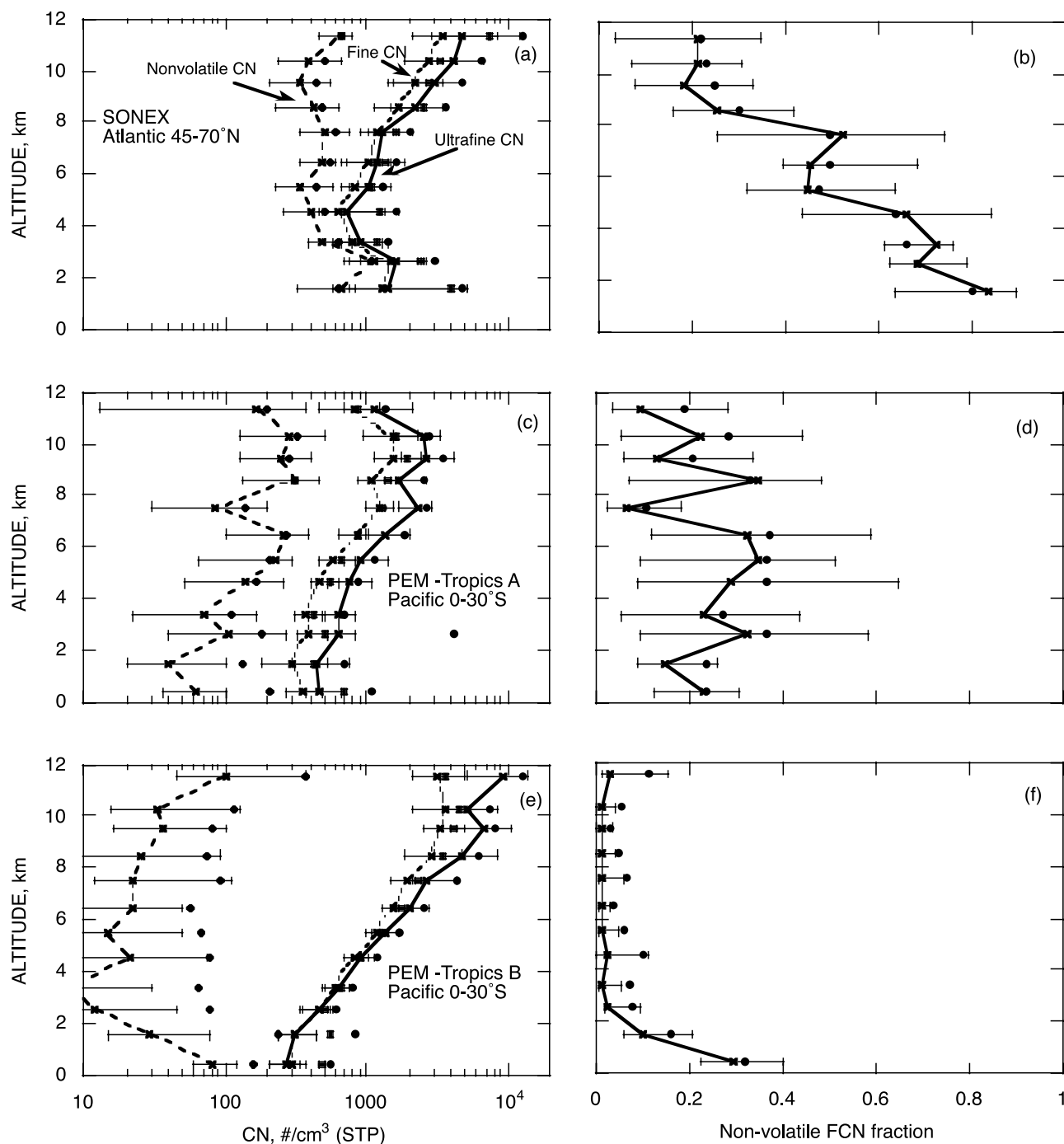
Figure 1. Vertical distribution of condensation nuclei in the remote troposphere: (a, b) data subset collected over the Atlantic (45–70°N; October/November 1997) during SONEX, (c, d) data subset collected over the Pacific (0–30°S) during PEM-Tropics A (September/October 1996), and (e, f) data collected over the Pacific (0–30°S) during PEM-Tropics B (March/April 1999). All particle data are normalized to 20°C and 1 atm. Ultrafine counts (UFCN) are for particle >8 nm diameter, and fine counts (FCN) are for diameter >17 nm. Nonvolatile FCN fraction is defined as NV-FCN/ total FCN. Mean (solid circles), median (crosses), and 25 percentile and 75% are shown. Drawn profiles are connecting medians.

[9] Using data from PEM-Tropics B, Figure 5 shows average vertical profiles of nonvolatile FCN and CO for three separate missions: Mission 19 (Tahiti to Easter Island), Mission 21 (Easter Island to Costa Rica), and Mission 22 (Costa Rica to southern California) [see *Raper et al.*, 2001].

Trajectory analysis [*Fuelberg et al.*, 2001] showed that air masses during these missions progressively moved from the clean environment of the tropical South Pacific Ocean (Mission 19) to areas impacted by pollution from Central America (Mission 21) and the North American mainland
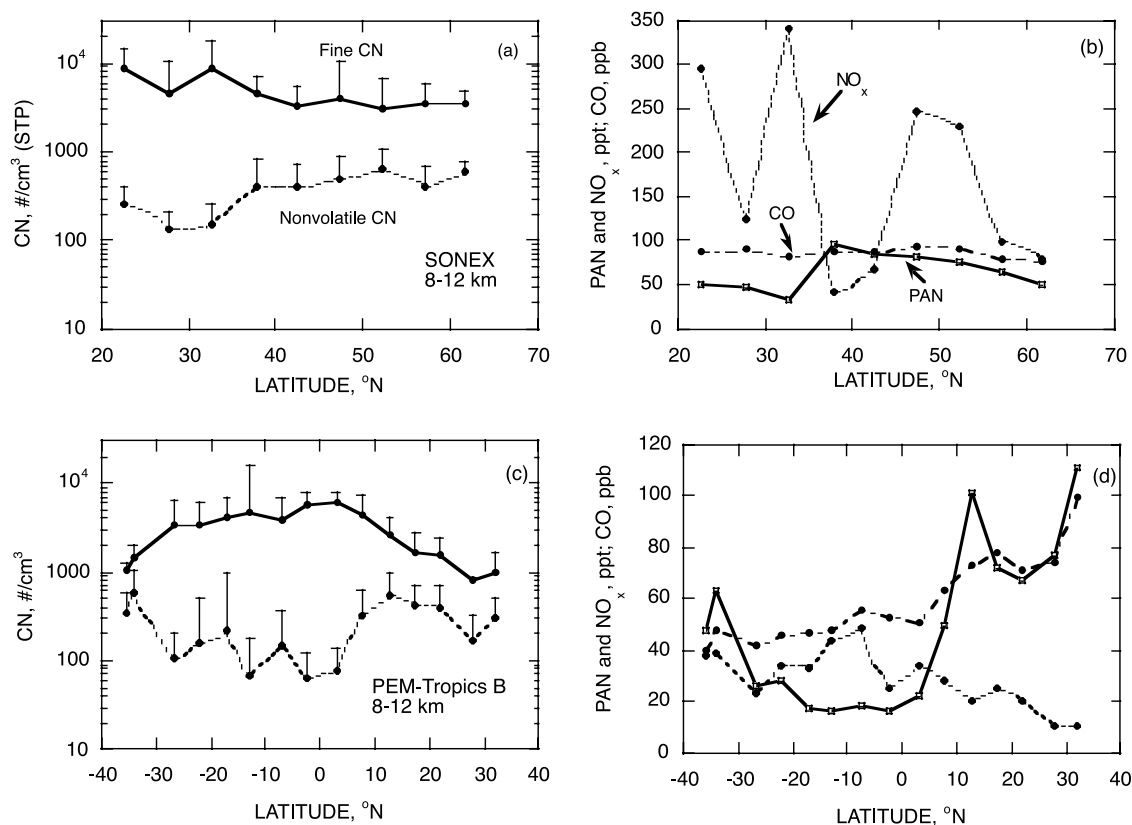
Figure 2. Latitudinal distribution of total and nonvolatile fine CN and selected gaseous chemicals (PAN, CO, and NO_x) in the upper troposphere (8–12 km) during (a, b) SONEX and (c, d) PEM-Tropics B. For clarity, error bars in Figures 2a and 2c are shown as $+1\sigma$.

(Mission 22). The response of nonvolatile FCN and CO is nearly identical at all altitudes. Other indicators of anthropogenic pollution (PAN, C_2H_2, SO_4^-, NO_3^-) behaved similarly. A clear commonality of nonvolatile particle with pollution sources is indicated. Thus it appears that nonvolatile particles in the UT were substantially of anthropogenic origin.

[10] The data in Figure 4 also suggest that the highest number densities of volatile fine particles are associated with the lowest nonvolatile particle concentrations and, in general, the least polluted conditions (low CO and PAN). Thus volatile particles in the UT do not appear to be strongly associated with anthropogenic pollution. This is consistent with the current view that these volatile particles are formed from processes involving gas to particle conversion and are most persistent when other surfaces are not available to scavenge the nucleating precursors (e.g., H_2SO_4). Gas to particle conversion of SO_2 to sulfuric acid via reaction with OH radicals is thought to be a key process for sulfuric acid formation [*Lovejoy et al.*, 1996; *Clarke et al.*, 1999; *Davis et al.*, 1999]. Binary nucleation involving the sulfuric acid-water system could be a principle process in UT particle formation [*Brock et al.*, 1995; *Hamill et al.*, 1997]. It also is possible that organic compounds (oxygenates, heavy alkanes) are involved in these processes [*Murphy et al.*, 1998], although their role in the UT has not been studied. While SO_2 is a common anthropogenic pollutant, a significant tropical source comes from the photochemical oxidation of dimethyl sulfide (DMS), known to be of

natural oceanic origin [*Berresheim et al.*, 1995]. Under normal circumstances this oxidation takes place in the lower troposphere. However, the DMS/SO_2 system can be rapidly transported to the UT during periods of active convection that are common in the tropics.

[11] Figure 6 plots the latitudinal distribution of DMS in the mixed layer (0–4 km) along with the UT concentration of ultrafine CN measured over the Pacific during austral spring and fall. The corresponding fine CN data are shown in Figure 2. A tropical DMS source is clearly evident with comparable DMS levels during spring and fall. Mean DMS levels in the UT were near the lower limit of detection (<2 ppt) although sporadic excursions were observed. Unlike other indicators of pollution (Figure 3), levels of UT SO_2 during PEM Tropics B ($\approx$25 ppt) were only slightly higher than in PEM Tropics A ($\approx$20 ppt). A climatological analysis indicated that convective activity in this tropical Pacific region during the austral spring (PEM-Tropics B) and fall (PEM-Tropics A) is comparable in intensity. This is further supported by the nearly identical vertical distribution of surface tracers (e. g., DMS, CH_3I) during PEM-Tropics A and B. Nucleation rates are a strong function of temperature and relative humidity [*Hamill et al.*, 1997]. During the cleanest periods most associated with particle formation, UT mean temperatures ($\approx$235 K) and relative humidities ($\approx$30%) during the two missions were also very similar. We surmise that the formation rates of fine particles during PEM-Tropics A and PEM-Tropics B were similar. The major reason for the large differences in
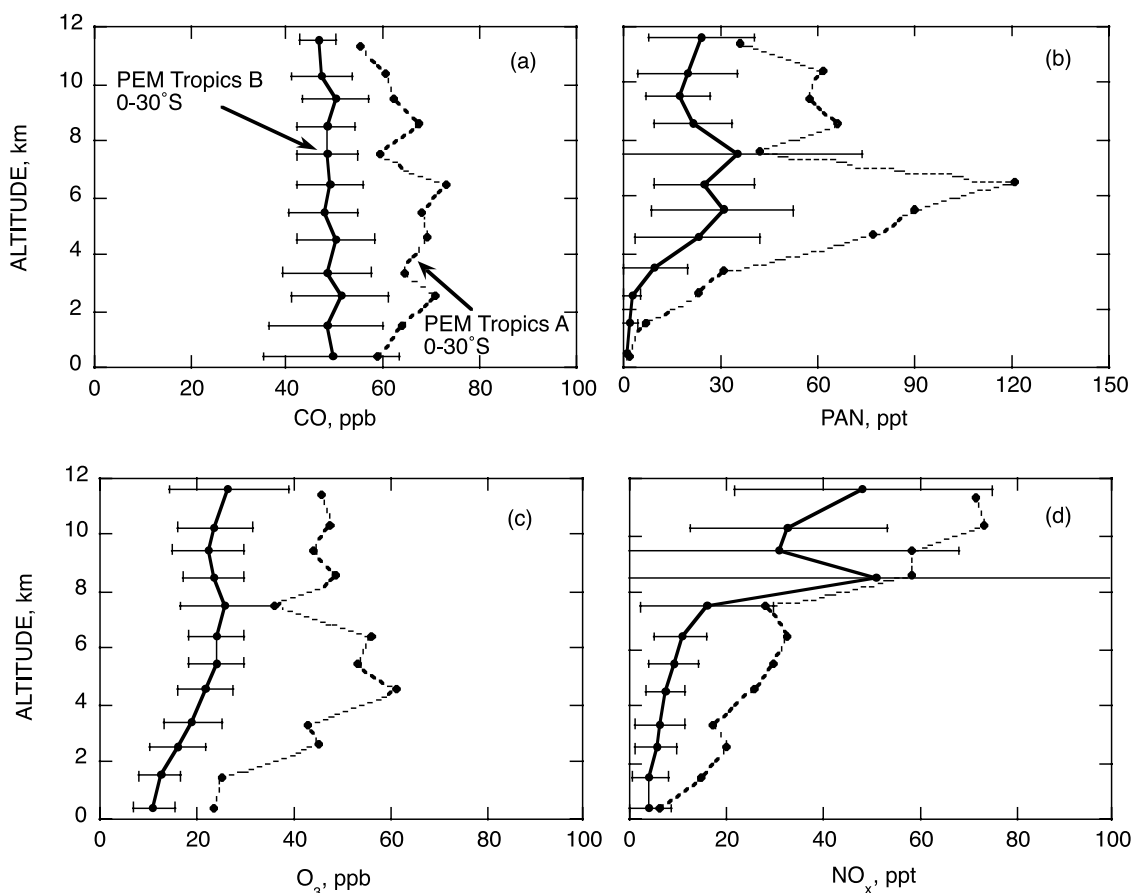
Figure 3. A comparison of the tropospheric mixing ratios of selected trace chemicals in the South Pacific (0–30°S) during PEM-Tropics B and PEM-Tropics A. For clarity, standard deviations for only PEM-Tropics B data are shown.

measured CN abundances likely arose from differences in loss rates. However, the possibility that other processes such as the direct scavenging of the gaseous precursor resulted in unequal formation rates cannot be ruled out.

[12] Results in Figures 1 and 2 indicate that the marine environment of the tropical UT is a large-scale source region of fine particles that are predominantly volatile. It is also evident from Figure 4 that the highest number densities of CN generally coexist with low nonvolatile FCN counts and minimum anthropogenic pollution as indicated by CO and PAN. The high levels of NO_x in the subtropical troposphere (Figures 2b and 2d) in both the Atlantic and the Pacific are almost certainly associated with the lightning source of NO_x in this region [*Levy and Moxim*, 1996]. We infer from Figures 1, 2, and 4 that the presence of nonvolatile particles tends to suppress the number densities of fine and ultrafine particles. In Figure 7 we plot mean volatile FCN number densities in the UT as a function of nonvolatile FCN using data from all three missions. These data can be conveniently fit to a power law curve, showing an inverse relation to be present. It is evident that volatile and nonvolatile particles do not favorably coexist and are anticorrelated. Increasing future emissions of nonvolatile particle from fossil fuel and biomass combustion should further reduce fine particle abundance in the UT. The degree to which these nonvolatile particles can absorb solar radi-

ation and provide nuclei for cloud formation is not known at this time.

[13] The inverse relationship between the number of volatile particles and the number of nonvolatile particles illustrated in Figure 7 led us to investigate its possible reasons. A number of possible mechanisms for the removal of volatile particles have been considered [*Fitzgerald et al.*, 1998]. Two likely scenarios are that (1) nonvolatile particles provide an efficient process for the removal of previously formed volatile particles via coagulation and (2) nonvolatile particles directly scavenge precursor gases (e.g., H_2SO_4) thereby suppress nucleation enough to prevent new particle formation. We tested scenario 1 by using a simple microphysical model in which the only process affecting the particles was coagulation [*Hamill et al.*, 1997]. Typically, our simulations began with initial conditions of 10^5 particles with a diameter of 4 nm and between 100 and 1500 particles with a diameter of 100–300 nm. We assumed that the small particles are volatile, while the large particles are nonvolatile, although this property of the particles has nothing to do with their coagulation characteristics. The assumption that the nonvolatile particles generally are larger is reasonable under at least some conditions; however, much smaller aerosol particles (e.g., soot) almost certainly are present [*Slinn*, 1975; *Warneck*, 2000]. We found that for coagulation times of ~3
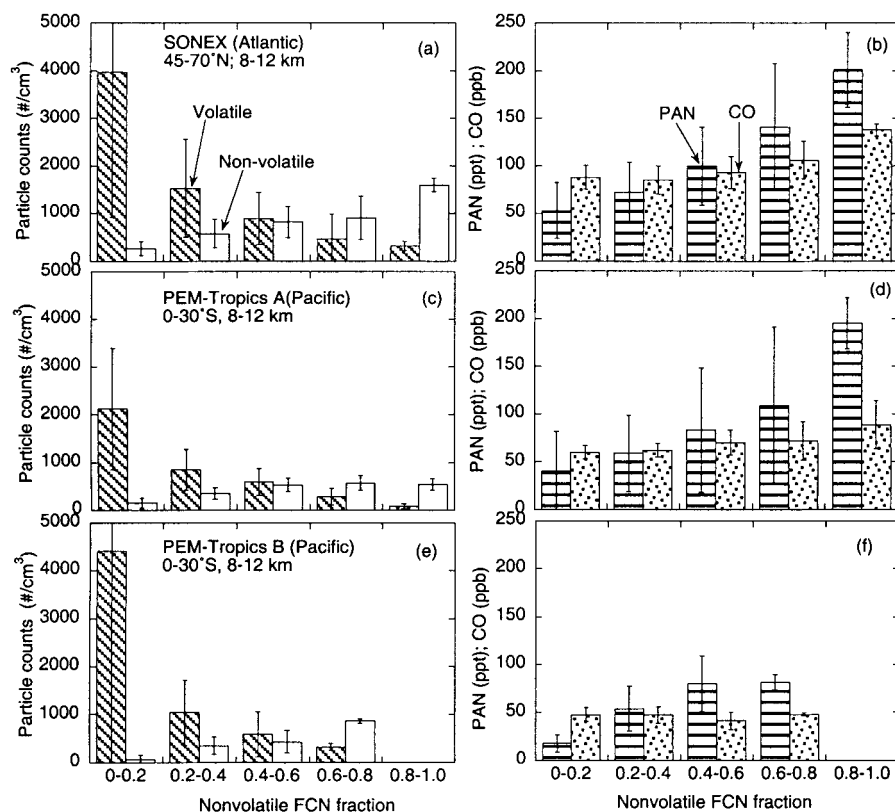
Figure 4. Variation of volatile and nonvolatile fine CN, PAN, and CO as a function of nonvolatile FCN fraction from data as in Figure 1. Error bars represent $\pm 1\sigma$.

days it was possible to develop a relationship similar to that in Figure 7. There are many unknown variables in this calculation, including the number of small (volatile) particles, the initial size distribution, the coagulation time, etc.

Nevertheless, it appears that coagulation with larger particles, presumably from pollution sources, contributes to the observed inverse relationship and offers one possible mechanism.
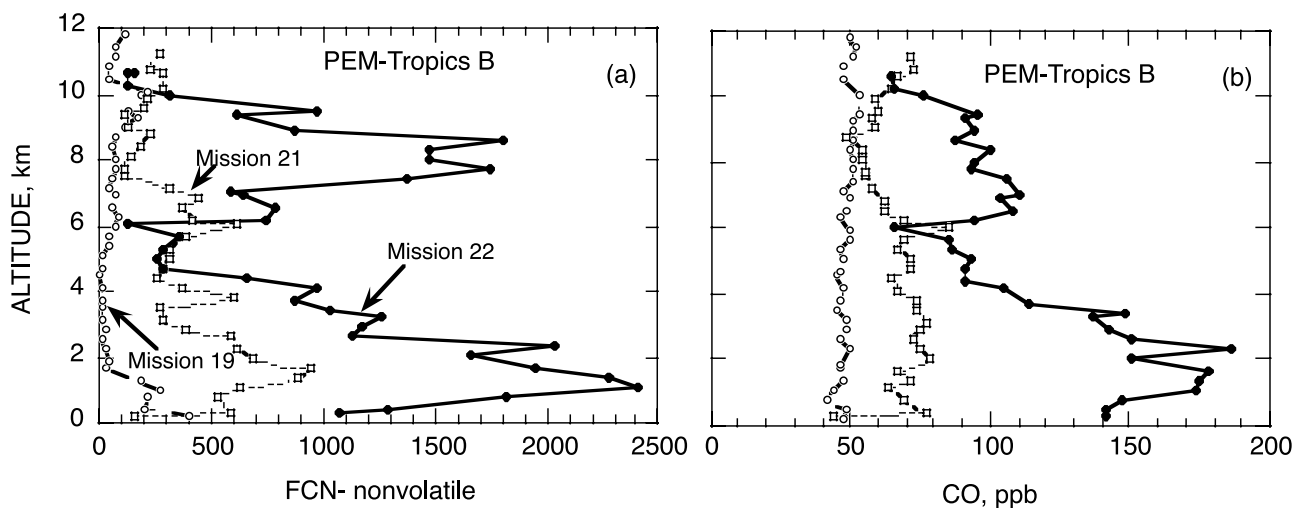

Figure 5. A comparison of the distribution of (a) nonvolatile fine CN (cm^{-3}) and (b) CO during progressive pollution events encountered over the Pacific in PEM-Tropics B. Mission 19 (13 April 1999), Tahiti to Easter Island; Mission 21 (17 April 1999), Easter Island to Costa Rica; and Mission 22 (18 April 1999), Costa Rica to southern California. More details on flight profiles are available from *Raper et al.* [2001].
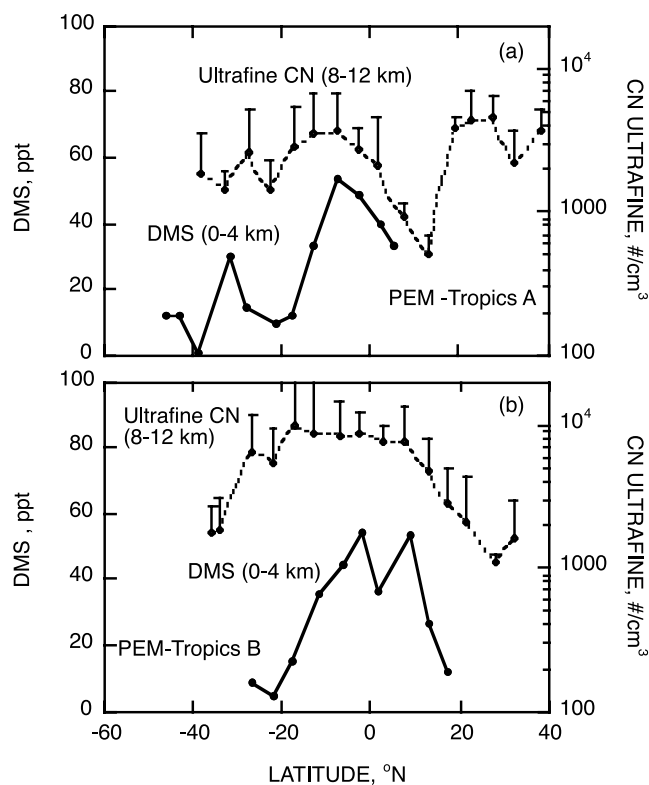
Figure 6. A comparison of lower tropospheric (0–4 km) DMS mixing ratios with upper tropospheric ultrafine CN counts over the Pacific during (a) PEM-Tropics A and (b) PEM-Tropics B.

[14] We also tested scenario 2 by assuming typical UT conditions found in PEM-West B (SO_2, 50 ppt; OH, 10^6 molecules cm^{-3}; T, 230 K) to see if the rate of SO_2 oxidation could sustain a H_2SO_4 concentration of $\approx 10^6$ molecules cm^{-3}, required for nucleation in the UT, in the presence of scavenging by large particles. These calculations employing reasonable sticking coefficients (0.5–1.0) showed the rate of H_2SO_4 formation ($\approx 10^3$ molecules cm^{-3} s^{-1}) was nearly 100 times slower in comparison to the calculated scavenging rates. Under such conditions, nucleation could indeed stop and new particle formation may not take place to any significant degree. This is often the case in the boundary layer where new particle formation is greatly suppressed. We speculate that a combination of both these processes plays a role in defining the shape of the anticorrelation observed in Figure 7.

[15] The abundance of fine aerosol is a balance between the production and destruction rates. While the UT tropical CN appear to be predominantly naturally produced, other sources of CN certainly are present. A comparison of FCN counts at similar latitudes (25–30°N; Figures 2a and 2c) suggests that FCN abundance is larger over the subtropical North Atlantic compared to the subtropical eastern Pacific. This could be due in part to the slightly lower NV-FCN number densities. We also note that during the cleanest conditions (NV-FCN < 0.2), most suited for particle formation, the UT over the North Atlantic during SONEX, in general, was colder and more humid (226 K; 60%) than the tropical Southern Pacific during PEM Tropics A or B (235 K; 30%). Simple microphysical calculations initiated with 10^7 molecules cm^{-3} of sulfuric acid show that
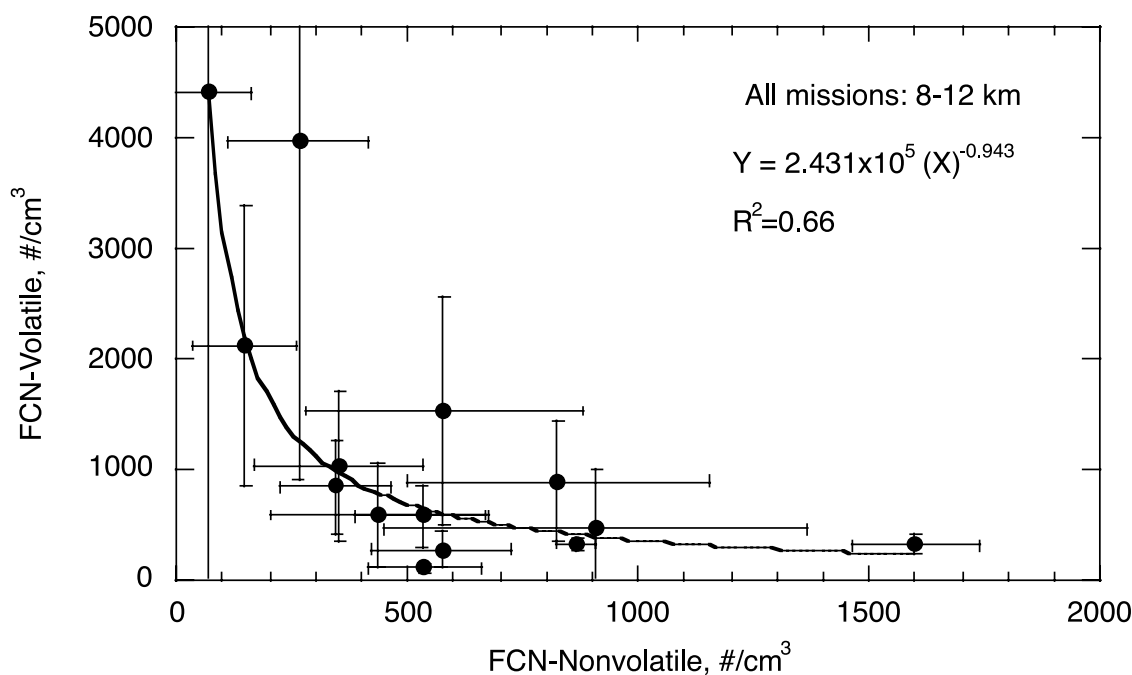

Figure 7. Relationship between the mean volatile and nonvolatile particle counts in the upper troposphere (8–12 km). The data are from SONEX, PEM-Tropics A, and PEM-Tropics B and represent means within the individual volatility bins plotted in Figure 4. Error bars represent $\pm 1\sigma$.
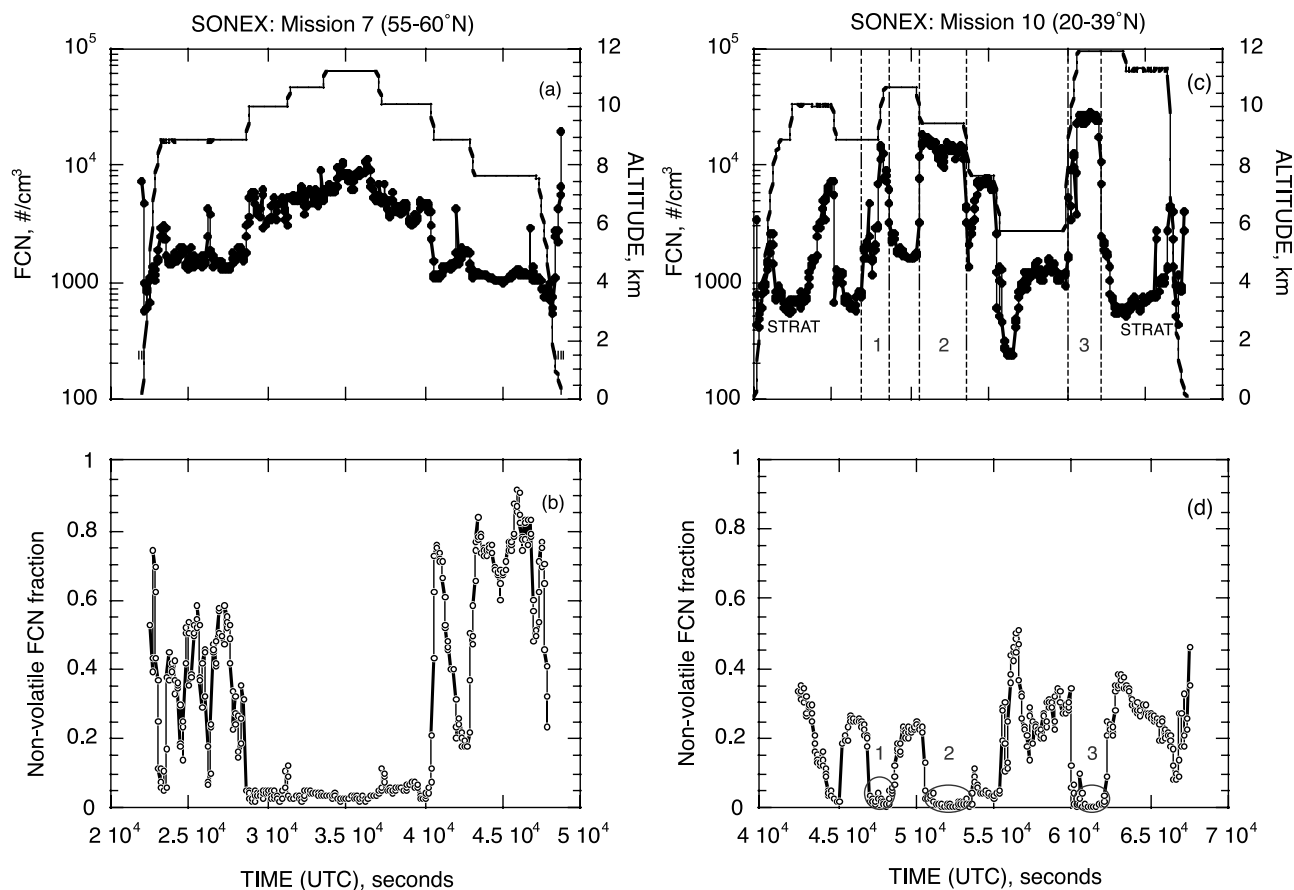
Figure 8. Variation of fine particle distribution and nonvolatile FCN fraction during two missions over the Atlantic during SONEX. Mission 7, 23 October 1997; Mission 10, 29 October 1997. More details on flight profiles are available from *Singh et al.* [1999].

particle formation rates during SONEX may have been $10^3 - 10^4$ times those during the Pacific missions. Thus the physical environment during SONEX may have been far more conducive to particle formation.

3.2. Transport of Clean and Polluted Air

[16] An important source of CN in the UT over the North Atlantic was due to commercial aircraft emissions [*Singh et al.*, 1999; *Ferry et al.*, 1999; *Anderson et al.*, 1999, *Schumann et al.*, 2000]. Figure 8 shows the distribution of FCN number density during two selected missions over the North Atlantic. Missions 7 (23 October 1997) and 10 (29 October 1997) are selected because they indicate two distinct sources of fine particles in the Atlantic UT. Mission 7 occurred within the North Atlantic Flight Corridor Organized Track System between 55 and 60°N where commercial air traffic was known to be heaviest. Figure 8a shows the dramatic increase in FCN within this corridor at 10–12 km. This increase consists of particles that are almost completely (>95%) volatile (Figure 8b). Using the time periods, geographic locations, and altitudes as receptor points, 5-day backward trajectory analysis showed that these air parcels had remained in the UT at midlatitudes. A source associated with emissions from commercial aircraft, along with the very cold temperatures encountered by the air parcels, provides the most satisfactory explanation.

Additional sources of CN in the UT came from episodes involving the rapid wet convection of polluted air from continental areas [*Wang et al.*, 2000]. These episodes were nearly always associated with lightning. It could not be determined whether lightning itself was responsible for particle formation.

[17] Fine particles sometimes were transported to the northern midlatitudes from sources in the tropical troposphere. Figures 8c and 8d show the DC-8 altitude profile, the fine CN number density, and the corresponding volatility during SONEX Mission 10. This flight was planned to be away from the aircraft corridor emissions. As is clear from Figure 8c, large variations in FCN were observed along the flight profile. Two periods when stratospheric air was sampled are labeled. Outstanding are three periods of high FCN counts exceeding 10^4 cm^{-3} (numbered in Figures 8c and 8d) observed at altitudes from 9 to 12 km that consisted almost completely of volatile particles. Using the DC-8 aircraft locations as receptor points, 5-day backward trajectories were obtained to examine the origin of the aerosol for the three cases [*Fuelberg et al.*, 2000]. Selected trajectories for the three cases are shown in Figure 9. It is evident that the high FCN containing air parcels were transported to the receptor areas over the North Atlantic all the way from the middle and upper troposphere of the tropical eastern Pacific to the Gulf of
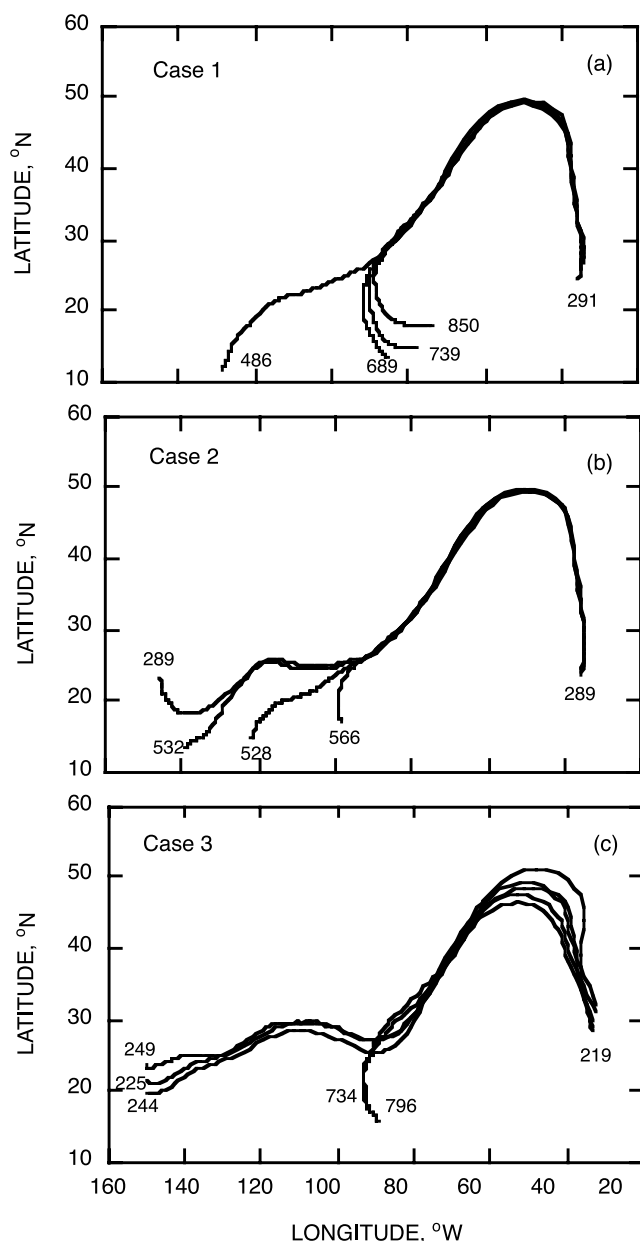
Figure 9. Five-day back trajectories representing sampling positions 1, 2, and 3 shown in Figure 8c. The numbers shown are pressure levels in millibars (mbar).

Mexico. These trajectories further show that this air had been much farther north in the UT prior to sampling. Simultaneous measurements of select tracers (e.g., CO, C_2H_2) further confirmed that the air parcels were of pristine tropical origin. Trajectories associated with the high nonvolatile fraction generally came from northwesterly directions. *Grant et al.* [2000] have also reported the presence of distinct periods over the eastern United States when large number densities of highly volatile FCN were transported from the tropical Pacific Ocean. These analyses suggest that periodic observations of large volatile aerosol number densities at mid to high latitudes can result from direct transport from their natural tropical/subtropical source region. In short, the situation over the

North Atlantic during SONEX was highly complex, with multiple sources superimposed on enhanced formation and destruction rates.

4. Conclusions

[18] Analyses of data from three recent airborne missions show that large number densities of highly volatile nuclei (CN) are present in the upper troposphere. The tropical/subtropical large-scale marine environments of the upper troposphere seem to contain the largest number of these particles. These particles are most abundant in the cleanest regions of the remote troposphere and appear to have a natural source. In selected regions and under certain conditions, rapid uplift of pollution and emissions from aircraft can also make a significant contribution to these number densities. Through long-distance horizontal transport, large numbers of volatile CN from the tropical/subtropical oceans can also be found in the middle and upper troposphere at mid and high latitudes. Nonvolatile particles are present in the upper troposphere as well. Their dominant source appears to be continental pollution associated with fossil fuel and biomass combustion. Data suggest that the presence of large nonvolatile particles causes volatile particles to be rapidly removed or not be formed in the first place. However, the mechanisms of the sources and sinks of these particles and their role in atmospheric chemistry and radiation are presently not well understood.

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